Field-induced magnetic ordering in $TlCuCl_3$: Lattice deformation and features of first-order transition.

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Abstract

We present NMR and strain-gauge study of a zigzag chain spin-gap material $TlCuCl_3$ which undergoes a field-induced long-range magnetic ordering transition regarded as a magnon Bose-Einstein Condensation (BEC). Deformation of the crystal lattice at the magnetic transition is inferred from the temperature dependence of the quadrupole shift of Cl and direct strain-gauge measurements, implying strong spin-phonon coupling. A λ-like singularity in spin-lattice relaxation rate at the transition reveals enhancement of electron spin fluctuations as precursor to transverse Néel order.

Key words: NMR, quantum spin chains, Bose-Einstein Condensation, phase transitions

1. Introduction

$TlCuCl_3$ belongs to the class of quasi one-dimensional (1D) spin liquids having non-magnetic singlet ground state separated from the first excited triplet by an energy gap Δ. Once the applied magnetic field is sufficient to destroy the gap ($H > H_{\rm g} = \Delta/g\mu_{\rm B}$) the system becomes partially polarized, and weak interactions between 1D segments can induce a long-range order (LRO). Depending on the nature and values of the exchange couplings, the properties of the field-induced gapless state are quite individual to each material.

The structure of $TlCuCl_3$ is monoclinic $P2_1/c$. Cu_2Cl_6 dimers are stacked in double chains along the a-axis and separated by Tl^+ ions in the bc plane. Intradimer AF interactions between spin-1/2 Cu^{2+} ions provide singlet ground state with $\Delta/k_{\rm B} \approx 7.5$ K [1]. However, 3D interactions are also quite strong due to sizable interdimer couplings [2]. The field-induced gapless state in $TlCuCl_3$ is resolved into a commensurate 3D transverse Néel order, considered as a Bose-Einstein condensate (BEC) of dilute magnons [3]. One of the signatures of the BEC is the power-law temperature dependence of the transition field, $g/2(H_{\rm LRO}(T) - H_{\rm g}) \propto T^{\phi}$, with generally anisotropic g-factor. BEC predicts ϕ=3/2. Experimentally however, $\phi = 2.1 \pm 0.1$ [1].

The magnon BEC is a unique phenomena deserving a careful study on the local level, which is a common objective of NMR. Besides, the existing discrepancy between the theoretical and experimental critical exponent has often been attributed to strong spin-phonon coupling, which needs to be experimentally checked. In this proceeding we report NMR and strain gauge studies of the transition of $TlCuCl_3$ to the field-induced magnetic long-range ordered state.

The sample preparation technique and experimental procedures have been described earlier [1,4]. All the measurements have been done in $H \perp (10\bar{2})$ configuration.

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2. Results and Discussion.

Static electronic properties (static magnetization and charge distribution) are borne in NMR lineshapes and peak positions. In our recent study [4] we have found a strong response of the crystal lattice to the magnetic ordering transition. This has been inferred from the abrupt 10% change of the quadrupolar shift of Cl at the LRO transition temperature, $T_{\rm LRO}$. Direct strain-gauge measurements have shown sizable changes of the crystal size at the ordering transition. The relative b-axis strain, $\Delta b/b$, was found negative and roughly linear in field, $(\Delta b/b)/H \propto -3.7\times10^{-6}{\rm T}^{-1}$. The a-axis strain is positive and the order of magnitude smaller than $\Delta b/b$. The observed involvement of the crystal lattice into the magnetic ordering transition evidences strong spin-phonon coupling in $TlCuCl_3$.

Scrupulous examination of NMR spectra in the vicinity of the transition has shown that the spectrum shape changes discontinuously, and that spectral features of both the ordered and the disordered phases coexist in 0.2 K×0.2 T region around the phase boundary. Taken together, these effects are quite indicative of the first-order transition, which is rather exotic for a purely magnetic system. At the same time, the observed involvement of the lattice could be a more common trigger for the first-order transition.

Low-frequency electron spin dynamics is probed by the nuclear spin-lattice relaxation rate, $1/T_1$. Fig. 1 represents the temperature dependence of ^{63}Cu $1/T_1$, measured in fields 4.8, 5.2, and 8.6 Tesla, i.e. below, at, and above $(g_\perp/2)H_{\rm g}$ ($H \perp (10\bar{2})$). At high temperatures $1/T_1$ goes as T^3 due to the spin gap, regardless of the field. Below 15 K the T-dependence is field-dependent. In 8.6 T, $1/T_1$ is peaked around 5.6 K which is $T_{\rm LRO}$ at this field. At $H = (g_\perp/2)H_{\rm g}$=5.2 T, $1/T_1$ is nearly constant at low temperatures. The low-temperature T-dependence in 4.8 T is linear. The field dependence of the relaxation rate measured at 1.5 K is shown in the insert in Fig. 1. The peak in $1/T_1$ around the phase boundary ($H_{\rm LRO}$(1.5 K)=5.35 T) is also present here.

The source for $1/T_1$ is transverse hyperfine field fluctuating at the NMR frequency. Therefore, the peak in $1/T_1$ at the phase boundary is evidently due to formation of the local hyperfine staggered field at Cu site $^{\rm Cu}h_\perp$, proportional to the transverse staggered magnetization component $M_\perp$. Existence of $M_\perp$ in the LRO state which signifies the transverse Néel order, has been detected in neutron scattering [5] and NMR [4]. $1/T_1$ shows however that the fluctuating component of $M_\perp$ arises in a finite region outside the ordered phase *prior* to the static $M_\perp$ which only shows up below the transition. Upon deeper penetration into the long-range ordered state, the fluctuations of $M_\perp$ freeze and $1/T_1$ diminishes, again as T^3.

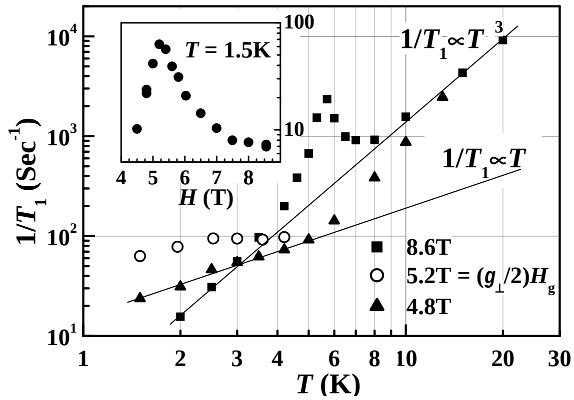

Fig. 1. ^{63}Cu spin-lattice relaxation rate. Main panel: Temperature dependence in three different fields. Insert: Field dependence at 1.5 K.

The T-dependence of $1/T_1$ around the phase transition in $TlCuCl_3$ is drastically different from that in spin-1/2 AF ladder $Cu_2(C_5H_{12}N_2)_2Cl_4$ [6], where proton $1/T_1$ turns down well above the transition temperature without any significant peak at the transition.

The weak T-dependence of $1/T_1$ at 4.8 and 5.2 T at low temperatures is indicative of quantum critical regime, similarly to $Cu_2(C_5H_{12}N_2)_2Cl_4$.

Summarizing, we have detected a strong response of the crystal lattice to establishment of the magnetic long-range order, which evidences strong spin-phonon coupling. Details of the NMR spectra in the close vicinity of the transition imply the first-order transition. The nuclear spin-lattice relaxation is strongly peaked around the phase boundary due to fluctuations of the transverse staggered magnetization component.

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